Review on gas-liquid mixing analysis in multiscale stirred vessel using CFD

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Abstract

This review aims to establish common approaches and equations used in computational fluid dynamics (CFD) analysis for gas-liquid mixing operations and investigate their strengths and weaknesses. The review concluded that with a sufficient computing strength, Eulerian-Lagrangian approaches can simulate detailed flow structures for dispersed multiphase flow with high spatial resolution. Turbulence is an important factor in fluid dynamics, and literature confirmed that k-ε is the most widely used turbulence model. However, it suffers from some inherent shortcomings that stemmed from the assumption of isotropy of turbulence and homogenous mixing, which is suitable for very high Reynolds number in unbaflled stirred vessels. In CFD simulations for gas-liquid systems in stirred vessels, bubble size distribution is the most important parameter; hence, different techniques for formulation of bubble size equations have been investigated. These techniques involve source and sink terms for coalescence or breakup and provide a framework in which the population balance method together with the coalescence and breakup models can be unified into three-dimensional CFD calculations. Different discretization schemes and solution algorithms were also reviewed to confirm that third-order solutions provide the least erroneous simulation results.

Keywords: computational fluid dynamics (CFD); frame grid; multifluid models; population balances; stirred vessel.

1. Introduction

Stirred vessels are widely used in the chemical, biochemical, mineral, pharmaceutical, and biotechnological industries to perform chemical operations such as halogenation, hydrogenation, oxidation, crystallization, polymerization, and biological fermentation (Van den Akker 2006, Buffo et al. 2012). Generally, mixing affects around 25% of all process industry operations (Yeoh et al. 2004). The primary aim of mixing is to increase the interfacial mass transfer rate in these vessels, which is related to the interfacial area between phases, volume fraction hold-up, bubble or droplet or particle size distribution, dynamic equilibrium between coalescence and breakage rate, turbulence level, fluid properties of dispersed and continuous phases, and mechanical mixing parameters such as impeller size and model, vessel size, and flow velocity (Bouuyatiotis and Thonton 1967). As it is impossible to cover all the mixing process parameters experimentally, computational fluid dynamics (CFD) has created opportunities to visualize the mixing phenomena (Haresh et al. 2010). CFD is able to predict fluid flows, chemical reaction rates, mass and heat transfer rates, and other occurrences by solving a set of appropriate mathematical equations (Ding et al. 2010). CFD also provides useful information for regions with intense or mild turbulence zones, Reynolds stresses, vortex structures, circulation patterns, flow behavior, and many other parameters (Haresh et al. 2010). Notwithstanding its potential, fundamental knowledge in multiphase flow simulations is still lacking because of its complexities, particularly in stirred vessels that have movable pieces. For example, CFD is still undeveloped to assess the non-homogeneity impact and phase property changes in stirred polymerization reactors (Haresh et al. 2010). Although review papers on experimental methods and geometrical parameters are available in literature (Mavros 2001, Ascanio et al. 2004), a complete review on CFD applications and mixing models in reactors is not yet available (Van den Akker 2006, Ochieng et al. 2009), which is the focus of this review.

This review provides an insight into different approaches used in simulating multiphase flow and different terms of momentum balances applied in gas-liquid systems in stirred vessels. The first term is momentum and stress transfer from the continuous to the dispersed phase, where different turbulence models have been investigated in this work. The second term is momentum transferred between the dispersed and the continuous phase, which depends on the force balance on each bubble and the dispersed phase distribution. Effects of drag and non-drag forces on bubbles were investigated based on force balance. The breakup and coalescence equations were investigated for distribution of dispersed phase. This review also identifies different methods available for solving population balance equations (PBEs). As an integrated solution for various equations is required, various framework, discretization schemes, and solution algorithms are also identified and analyzed. In addition, CFD simulation using mass transfer was also reviewed because results for these runs can also be analyzed from a mass transfer perspective.

2. General approaches

Two approaches are available for multifluid modeling, Eulerian-Eulerian (E-E, a two-fluid model) and Eulerian-Lagrangian (E-L). In the E-E multifluid method, the
continuous phase and the different dispersed phases are treated as a continuous interpenetrating media and described in terms of their volume fractions. The flow fields are solved for both phases, and the interactions between the phases are explained through the source terms (Buffo et al. 2012). The interfacial momentum transfer between liquid and gas in this model includes investigations on the influence of turbulent fluctuations on the effective momentum transfer, mass force, Basset force, lift force, and the drag force (Van Wachem and Almstedt 2003). However, the Eulerian approach suffers from numerical diffusion from the smearing of the gas fraction over the entire grid cell (Sokolichin et al. 1997). In the E-L approach, the continuous phase is treated in a Eulerian framework (using averaged equations), whereas particles are tracked through the flow in a Lagrangian manner (Ding et al. 2010). A force balance is solved for each particle, and each particle is individually tracked in the system (Ashraf Ali et al. 2008). The second Newtonian law is also solved for each individual particle (Ashraf Ali and Pushpavanam 2011), and a collision model is applied to handle particle encounters (Van Wachem and Almstedt 2003). Generally, E-L models simulate flow structures with higher spatial resolution compared with E-E models and show more benefits for dispersed multiphase flows. Other benefits include the following:

- Reactions occurring within or on surface of individual particles, and different transport processes can be modeled in a realistic way.
- Bubble coalescence and breakup, four-way momentum exchange, and bubble-induced turbulence can be rigorously modeled.
- The exchange of momentum between the operating phases due to drag, lift, and virtual mass through the interface can be modeled more accurately in comparison with E-E.
- Dispersed size distribution can be accounted without great difficulty (Buwa et al. 2006).
- Time-averaged properties could be predicted well using this viewpoint (Buwa et al. 2006).

However, the E-L approach faces three problems: false fluctuations of the continuous phase velocity result in sudden changes of dispersed phase local volume fraction; the amplitude of the false velocity fluctuation increases when a small grid is applied; as the volume fraction of dispersed phase increases, the interaction between the two phases will increase as well, which is not accounted for by this method due to the considerably high demand of computational cost (Ochieng and Onyango 2010); thus, this approach for a three-dimensional (3D) flow is more applicable in dilute systems (Yeoh et al. 2005).

In both approaches, there are two basic equations that should be solved, continuity and momentum balance, as given in eqs. (1) and (2), respectively:

\[
\frac{\partial}{\partial t}(\alpha_\ell \rho_\ell U_\ell) + \frac{\partial}{\partial x}(\alpha_\ell \rho_\ell u_i u_i) = \Gamma
\]  
\[
\frac{\partial}{\partial t}(\alpha_\ell \rho_\ell U_\ell) + \frac{\partial}{\partial x}(\alpha_\ell \rho_\ell u_i u_i) = - \alpha_\ell \frac{\partial P}{\partial x} + \alpha_\ell \rho_\ell g + \frac{\partial}{\partial x}T + \alpha_\ell F_{\text{H}}
\]

where the \( T \) is the stress tensor from the liquid and \( F_{\text{H}} \) is the momentum transferred through the force balance.

3. Stress tensor from the continuous phase

Eq. (3) expresses the turbulence treated through stress tensor terms \( (T_{ij}) \) in the momentum equation balance.

\[
\bar{T}_{ij} = \alpha_\ell (\mu_\ell + \mu_\omega) \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \frac{\partial u_k}{\partial x_k} \right) - \alpha_\ell \frac{\partial}{\partial x_j} (\rho_\ell u_i u_j) 
\]  

The term \( \rho_\ell u_i u_j \) introduces the Reynolds stresses.

\[
\rho_\ell u_i u_j = \frac{2}{3} \rho k \delta_{ij} + (\mu_\ell) \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) 
\]

A variety of turbulence models are available by coupling this term to other terms in the momentum balance equation, such as Reynolds averaged Navier-Stokes (RANS), large eddy simulation (LES), and the direct numerical simulation (DNS).

3.1. RANS model

The definition of kinetic energy of fluctuating motions is an aspect of concern in CFD modeling of stirred vessels, which is defined as the summation of the periodic component (due to nonrandom oscillations caused by the cyclic passage of the impeller blades) and the random component (due to turbulent eddies). RANS-based models depend on the random component of fluctuating motions to reduce computational effort by avoiding resolution of unsteady turbulent eddies directly (Montante et al. 2001).

RANS equations are divided into two categories: the Reynolds stress models (RSMs) and the eddy-viscosity model. The two-equation eddy-viscosity models are based on the assumption that an analogy exists between Reynolds stress and viscous stress and the turbulent flow is isotropic. Standard \( k-\epsilon \), renormalization group \( k-\epsilon \) (RNG), and realizable \( k-\epsilon \) are three different appearances of two equations, the \( k-\epsilon \) family model and the \( k-\omega \) models, where \( k \) is the turbulent kinetic energy, \( \epsilon \) is the energy dissipation rate, and \( \omega \) is the turbulent frequency. Table 1 shows the relevant equations in literature. Mixture, per phase, and dispersed are the three different extensions of the standard \( k-\epsilon \) turbulence model available for simulating turbulence in multiphase systems. This method is not adequate to capture the tracer concentration history, which is used to calculate the liquid-phase mixing time. In addition, anisotropy consideration and different time scales do not lead to a significant improvement over the standard \( k-\epsilon \) turbulence model (Kasat et al. 2008). Some improvements on the performance were achieved through RNG and realizable
Table 1 Summary of turbulence models.

<table>
<thead>
<tr>
<th>Model</th>
<th>$S_k$</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>RANS-based models</td>
<td>$\rho \left( C_{1s} \frac{\varepsilon}{k} C_{2s} \frac{\varepsilon}{k} \right)$</td>
<td>$C_{\rho_s} = 0.009$, $C_{\varepsilon_s} = 1.44$, $C_{\varepsilon_s} = 1.92$, $\sigma_{k,s} = 1$, $\sigma_{\varepsilon,s} = 1.314$</td>
</tr>
<tr>
<td>RNG (Rahimi and Parvareh 2005)</td>
<td>$\rho \left( C_{1s} \frac{\varepsilon}{k} C_{2s} \frac{\varepsilon}{k} \right)$</td>
<td>$\alpha = C_{\rho_s} \eta^{1-\eta/\eta_s}$, $C_{\rho,RNG} = 0.845$, $C_{\varepsilon,RNG} = 1.42$, $C_{\rho_s} = 1.68$, $\alpha_{k,s} = 0.719$, $\eta = 4.8$, $\beta = 0.012$, $\eta = E_k / \varepsilon$, $E_k = 2E y$, $E_y = 0.5 \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$</td>
</tr>
<tr>
<td>Realizable $k$-$\varepsilon$ model (Rahimi and Parvareh 2005)</td>
<td>$\rho \left( C_{1s} \frac{\varepsilon}{k} C_{2s} \frac{\varepsilon}{k} \right)$</td>
<td>$C_{\rho_s} = \max \left[ 0.43 \frac{\eta}{\eta_s + 5} \right]$, $C_{\varepsilon_s} = 0.09$, $C_{\varepsilon_s} = 1.9$, $\sigma_{k,s} = 1.2$, $\eta = E_k / \varepsilon$, $E_k = 2E y$, $E_y = 0.5 \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$</td>
</tr>
</tbody>
</table>

$k$-$\omega$

$$\frac{\partial \omega}{\partial t} + \frac{\partial (\omega u_1)}{\partial x_1} = \frac{\partial}{\partial x_1} \left[ \left( \nu + \frac{k}{\omega} \right) \frac{\partial \omega}{\partial x_1} \right] + \frac{\partial}{\partial x_2} \left[ \left( \nu + \frac{k}{\omega} \right) \frac{\partial \omega}{\partial x_2} \right]$$

$$\nu = \frac{k}{\omega}, \quad \tilde{\omega} = \max \left[ \omega, C_{\text{iso}} \left( \frac{2S_k S_{\omega}}{\beta} \right) \right]$$

$$\alpha = 0.52, \quad \beta = \beta_{ref}, \quad \beta_s = 0.0708, \quad \beta' = 0.09, \quad \sigma = 0.5$$

$$\sigma' = 0.6, \quad \gamma_s = \frac{1 + 85 \nu}{100 \nu}, \quad \chi = \frac{\Omega_k \Omega_{\omega}}{\beta \omega}$$

$$\sigma = 0 \text{ for } \frac{\partial k}{\partial x_1} \frac{\partial \omega}{\partial x_1} \leq 0, \quad \sigma = 0.125 \text{ for } \frac{\partial k}{\partial x_1} \frac{\partial \omega}{\partial x_1} > 0$$

RSM

$$\frac{\partial \sigma}{\partial t} + \frac{\partial \sigma u_i}{\partial x_i} = \left( \tau \frac{\partial u_i}{\partial x_i} + \tau_{ij} \frac{\partial u_j}{\partial x_i} \right)$$

$$\frac{\partial \tau_{ij}}{\partial t} \sigma = \frac{\partial}{\partial x_j} \left[ \left( \nu + \frac{k}{\omega} \right) \frac{\partial \tau_{ij}}{\partial x_j} \right]$$

$$\Pi_i = \left( P' \frac{\partial u_i'}{\partial x_j} \right) \left( \frac{\partial u_j'}{\partial x_j} \right)$$

$$C_{ij} = \frac{1}{\rho} \left( P' \frac{\partial u_i'}{\partial x_j} \right) \left( \frac{\partial u_j'}{\partial x_j} \right)$$

$$\sigma_i = 0.82 C_{ij}$$

LES

$$\frac{\partial u_i}{\partial t} + \frac{\partial u_i u_j}{\partial x_j} = -\frac{\partial P}{\partial x_j} + \frac{\partial}{\partial x_j} \left( \frac{\partial u_i}{\partial x_j} \right) + \frac{1}{\rho} \left( \frac{\partial u_i}{\partial x_j} \right)$$

$$\tau_{ij}^{RNG} = \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$$

$$\tau_{ij}^{LES} = \frac{1}{3} \tau_{ij}^{RNG} \delta_{ij} - 2 \tau_{ij}^{LES} S_{ij}$$

$$v_{LES}^2 = L_x \left[ S_{ij}^2 v_{LES}^2 \right]$$

$$S_{ij}^2 = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)$$

$$L_x = \min \left( C_{ij} \gamma_{ij}^{LES} \right)$$

models (Wang et al. 2010). The effect of small-scale turbulence is introduced in the RNG $k$-$\varepsilon$ model through a random forcing function in the Navier-Stokes equations. In this model, the effect of small-scale motion from the governing equations is removed by a modified viscosity and by expressing their effects in terms of larger-scale motion (Hjertager et al. 2002); thus, the RNG model gives better results in predicting the turbulent kinetic energy in the wall jet below the impeller compared with standard $k$-$\varepsilon$, which overpredicts it (Harris et al. 1996). The realizable $k$-$\varepsilon$ contains a new transport equation for the dissipation rate ($\varepsilon$) and a new formulation for the turbulent viscosity. $C_H$ (a constant in the definition of the turbulence length scale) is not a constant in this model and is instead a function of local strain rate and rotation of fluid. Furthermore, different source and sink terms are used in this model in the transport equation for eddy dissipation. Both the RNG and the realizable $k$-$\varepsilon$ models have indicated important improvements over the standard $k$-$\varepsilon$ model where the flow features include strong streamline curvature, rotation, and vortices (Rahimi and Parvareh 2005). Despite all these modifications,
the \( k-\epsilon \) turbulence model provides unrealistic results in several cases due to inaccurate turbulent kinetic energy distribution prediction especially in the regions close to the impeller, particularly due to the isotropic nature of the \( k-\epsilon \) turbulence models and the steadiness and transient nature of the flow (Zadghaffari et al. 2010, Coroneo et al. 2011) within these regions. Assumption of isotropic turbulence and homogeneity only produces accurate results for flows at high Reynolds numbers (Trivellato 2011). In this situation, it is assumed that the energy is injected into eddy motions on a scale in the order the size of eddies and then transferred to smaller length scales until it is dissipated at the smallest stable viscous scale (the Kolmogorov scale) (Trivellato 2011), but this assumption is not practical in real systems. Bujalski et al. (2002) reported the calculation of power number through the integration of turbulent energy dissipation rate over the whole vessel, which was about 20% lower than the experimental value. This problem became more severe in regions where the flow is highly anisotropic and in systems containing high solid loading (Ochieng and Onyango 2010); thus, the \( k-\epsilon \) model cannot give accurate results for the tangential velocity components and in describing curvilinear and highly swirling flows. The RSM is another turbulence model that solves the transport equations for all Reynolds stresses, represents the full effect of turbulence in the momentum equations, and removes the assumption of the isotropic eddy viscosity. However, RSM has shortcomings due to numerical difficulties and high computational effort and suffers from poor convergence behavior. Additionally, both transient and steady-state results using RSM have underpredicted turbulence kinetic energy (Montante et al. 2001, Murthy and Joshi 2008). Hence, a compromise algebraic stress model (ASM) was developed to overcome these problems.

In the ASM model, the convection and diffusion terms are derived from a local equilibrium condition, and the Reynolds differential equations are converted into a set of algebraic equations. However, there is no diffusion or damping factors in the equations, and the expressions are implicit for Reynolds stress components; hence, this model is not applicable strong and cannot perform for complex 3D flows. The explicit algebraic stress model (EASM) is another version of ASM that was developed using a tensor polynomial expansion theory. In this method, the Reynolds stresses was described as an explicit algebraic correlation of turbulence characteristic quantities, mean strain rate tensor, and rotation rate tensor. In comparison, the EASM produces numerical stability and lower computational effort. Additionally, the EASM also performs well in comparison with the DNS results as explained later in this work. Feng et al. (2012) conducted a simulation for complex 3D turbulent flows in a stirred vessel and suggested the EASM as a suitable alternative model to the standard \( k-\epsilon \) model for simulating anisotropic turbulent flows in agitated vessels. As \( k-\epsilon \) model equations cannot be integrated to the wall, \( k-\omega \) have been derived, where the turbulence energy dissipation rate is replaced by the turbulence eddy frequency, \( \omega \), allowing detailed behavior inside the boundary layer to be calculated and performs well near walls. However, it behaves poorly away from walls due to oversensitivity to free stream conditions. Shear stress transport (SST) (Menter 1994) is a hybrid model that combines the best features of \( k-\omega \) and \( k-\epsilon \) models with a turbulence production limiter in the calculation of eddy viscosity. Blending between the \( k-\omega \) model in the near-wall region and the \( k-\epsilon \) model in the free stream using a smooth function gives a marginal improvement in the region closer to the wall. However, it is still unable to resolve any details of unsteady turbulent structures in the bulk fluid region. The two methods fail to properly capture the chaotic and 3D nature of trailing vortices that form downstream of the impeller blades (Ochieng and Lewis 2006). A significant shortcoming of unsteady two-equation models is the inability to resolve any of the turbulence details directly due to excessive damping of turbulence (Menter 2009). A complete review on the models mentioned above was done by Joshi et al. (2011a).

### 3.2. LES model

Some authors reported poor CFD results due to turbulent kinetic energy (Jaworski et al. 2001, Aubin et al. 2004). Many authors attended to the LES method (as shown in Table 1), which was first published for a stirred vessel by Eggels (1996). It is worth mentioning that the Smagorinsky-Lilly model is an eddy-viscosity model, which is the most common model for predicting the dissipation rate in the LES model (Smagorinsky 1963). In LES, large scales eddies are resolved, and the small scales, which are isotropic in nature, are modeled using subgrid scale models. The major role of subgrid scale models is to provide proper dissipation for the energy transferred from the large scales to small scales eddies (Joshi et al. 2011a).

The LES has been shown to perform well in capturing the complexity of turbulent flow in a mixing reactor (Hartmann et al. 2004, Yeoh et al. 2004, Delafosse et al. 2008). It is able to overcome \( k-\epsilon \) limitations for investigating unsteady behavior in turbulent flow and provide details of flow field that cannot be obtained by RANS (Derkson et al. 1999). However, it is weak in cases where rate-controlling processes occur at smaller scales. As the dissipation rate cannot be resolved during LES simulation, it is estimated \textit{a posteriori} and strongly depends on the subgrid scale model used in the simulations (Delafosse et al. 2008). The LES is also not fine-tuned for quick process design validation and still requires rather intense CPU time. There has not been any validation for high-solid-concentrations slurries using the LES, although in contrast to RANS, it is still easier to recognize the part of fluctuations (Hartmann et al. 2004). Menter (1994) have developed a turbulence model called scale adaptive simulation-shear stress transport (SAS-SST), which is based on the idea of capturing as much of the turbulence field as possible and using RANS capabilities near walls and regions of “steady” flow (Hartmann et al. 2004, Honkanen et al. 2007). In this model, small-scale turbulences are dealt through Reynolds averaging, whereas larger-scale turbulences are resolved directly. This model has shown accurate prediction of the length and intensity of trailing vortices generated by impeller blades in a stirred vessel. However, it requires large numbers of modeled
revolutions to obtain good statistical averaging for calculating turbulence quantities (Singh et al. 2011). Further details on single and multiphase turbulent flow models are available in the reviews by Joshi et al. (2011a,b).

3.3. DNS model

The DNS is another option that resolves all time scales and turbulent length where basic equations are constructed with no turbulent and interface models. Navier-Stokes equations are directly integrated using a very fine grid. This method is not feasible for an industrial scale because it requires enormous amounts of grid cells, particularly for bubbly flows, because of the very short relaxation time of bubble motions in liquid in comparison with liquid drops and the deformability of the bubble surface. The large density ratio of bubbles to bulk liquid makes this approach more complicated (Kitagawa et al. 2001). Furthermore, it has some restrictions with small Reynolds and Schmitt number (Chakrabarti and Hill 1997), as an example, at the lower limit of a turbulent regime, it needs very fine grids in the critical flow regions (vicinity of the impeller) to fully and accurately resolve the flow (Hartmann et al. 2004), and it is also algebraically complex and numerically inefficient (Forney and Nafia 2000).

Although LES and DNS approaches are generally more accurate for turbulent fluid mixing (Hartmann et al. 2004, Singh et al. 2011), the RANS approach remains the most popular simulation method because it requires less computational effort and time. Although different specific conclusions have been drawn by various researchers in terms of local turbulent quantities as mentioned in Table 2, generally, comparison of RANS and LES simulations has led to the conclusion that the latter is more superior simply because more satisfactory agreement between measured and calculated data has been reported for the LES. In summary, the LES methodology has advantages compared with the RANS approach because the velocity fluctuations, Reynolds stresses, and turbulent kinetic energy are resolved down to the scale of the numerical grid. Also, the LES presents levels and structure of turbulent kinetic energy in impeller discharge flow better than the RANS simulation. The levels of energy predicted by the RANS simulation were higher than that predicted by the LES; this was also observed for the spreading rate of energy dissipation. In comparison, transient RANS simulation can accurately present flow fields although turbulent kinetic energy predictions are still poor, especially in discharge flow and impeller region where most mixing takes place. A detailed explanation is also available in the reviews by Joshi et al. (2011a,b).

4. Momentum transferred between dispersed and continuous phase

Another important term in the momentum balance equation is the momentum transferred between bubbles (as the dispersed) and the liquid (as the continuous) phase, which is defined in eq. (5). The value is dependent on the dispersed phase volume fraction ($\alpha$) (which will be explained in the next section) and the momentum transferred ($\dot{F}_m$):

$$\alpha \dot{F}_m = \left( \frac{1}{V_{cell}} \Delta t \right) \sum \left( F \right) \Delta t,$$

where $V$ is the volume of the cell, $\Delta t$ is the time step, $E$ is the cumulative dissipated energy, and the last term is the summation of the drag and nondrag forces. In addition, by solving the force balance on each bubble, the bubble velocities can be calculated using eq. (6):

$$m_b \frac{d\mathbf{u}_b}{dt} = F_c + F_p + F_D + F_L + F_{VM}$$

The left side of eq. (6) represents $m_b$ and $u_b$, which are bubble mass and velocity, respectively. The right side of the equation is the summation of all forces acting on the bubble, which is divided into two major groups: drag and nondrag forces.

4.1. Nondrag forces

In systems that include continuous and dispersed phases, the assessment of interactions between the operational phases and the effect of continuous phase properties on the dispersed phase in the CFD modeling can be introduced by force balance on bubbles (Scargiali et al. 2007). Apart from drag force, various nondrag forces affect the bubble’s motion when moving through a fluid and the motion is non-uniform. These nondrag forces result in interphase momentum exchange beside the drag force. They are Basset history force, lift force, and virtual mass force. Mass force is an inertial force caused by relative acceleration between the secondary phase and the primary phase (Khopkar and Tanguy 2008). Basset force is a viscous force caused by the development of a boundary layer around bubbles and relative acceleration. It is relevant only for unsteady flows. Lift force denotes the transverse force caused by rotational strain, velocity gradients, vorticities, and shear in the continuous phase flow field. Thus, in the bulk region of stirred vessels, lift force is much smaller than in the vicinity of the impeller (Lane et al. 2002).

4.2. Drag force

The interaction force between phases especially in aerated stirred vessels is mainly due to the drag force. The effects of virtual mass, turbulent dispersion, and lift are almost negligible despite a significant increase in convergence difficulties and computational expenses. For example, Scargiali et al. (2007) have reported an increase of overall gas holdup from 4.36% to 4.67% and from 4.36% to 4.60% by considering the effect of lift force and virtual mass, respectively. Similar results have been indicated by many other authors (Bakker et al. 1994, Lane et al. 2002, Kerdouss et al. 2006), but depending on the gas holdup and flow regime, the role of these forces can be important. Some authors have reported that on-drag forces are mainly responsible for lateral migration of bubbles and distribution of gas volume fraction in
Table 2 Comparison of LES and RANS in mixing vessels.

<table>
<thead>
<tr>
<th>Authors/year/system, impeller type, speed, T, D/T, C/T, N, W/T, Z/T, operating material, Re</th>
<th>Grid size, type, impeller motion model</th>
<th>Flow variable</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Yeoh et al. 2004, RT, 36.08 rps 100 mm, T/3, T/3, 4, T/10, T, water, 40,000</td>
<td>15,672, hexahedral (full), sliding/deforming, CFD code: in-house coding</td>
<td>( \langle u \rangle )</td>
<td>Purpose: To simulate turbulent flow RANS model: standard ( k-\varepsilon ) Notes: The global turbulent energy dissipation rate ( \varepsilon ) across the vessel volume was underpredicted by 45% with the RANS model; in contrast, ( \varepsilon ) was well predicted by the LES to within 15% of the measured value</td>
</tr>
<tr>
<td>Hartmann et al. 2004, RT, 2672, 150 mm, T/3, T/3, 6, T/10, T, silicon oil density, 1039 kg/m³, dynamic viscosity, 15.9 mPa s, 7300</td>
<td>228,096, hexahedral (full), SM, CFD code: CFX 5.5.1.</td>
<td>( \langle u \rangle )</td>
<td>Purpose: To compare URANS and LES RANS model: SST Notes: The LES calculations agree better with the experimental results than the RANS calculations</td>
</tr>
<tr>
<td>Yeoh et al. 2005, 100 mm, T/3, T/3, 4, T/10, T, neutrally buoyant tracer, –, 40,000</td>
<td>490,000</td>
<td>Mixing pattern, time trace of normalized concentration</td>
<td>Purpose: To characterize the mixing of an inert scalar and comparison of URANS and LES RANS model: standard ( k-\varepsilon ) Notes: There is a reasonable agreement with experimental, with an average deviation of 18% using the LES</td>
</tr>
<tr>
<td>Jahoda et al. 2007, six-blade 458, pitched blade turbine, a standard RT, 0.29 m, T/3, T/3, 2, T/10, 2T, tap water, ( 4.66 \times 10^4 )</td>
<td>61,500 for one impeller, 1,230,000 for two impeller hexahedral (full), SM, MRF, CFD code: FLUENT 6.2</td>
<td>( \langle u \rangle )</td>
<td>Purpose: To predict liquid homogenization RANS model: standard ( k-\varepsilon ) Notes: The LES approach described the real flow with best agreement</td>
</tr>
<tr>
<td>Murthy and Joshi 2008, DT, standard PBTD60, 45, and 30, HF, 4.5 rps, 0.3 m, T/3, 0.082 m, 4, T/10, T, water, –</td>
<td>575,000 for RSM, 1,275,567 for LES, hexahedral, SM, CFD code: FLUENT 6.2</td>
<td>( \langle u \rangle )</td>
<td>Purpose: To investigate URANS and LES RANS model: standard ( k-\varepsilon ), (RSTM) Notes: All the three turbulent models predicted the power number (NP) well, but by integration of local turbulent energy, the dissipation rate value obtained from the RANS-based models was underpredicted in the range of 20–25%</td>
</tr>
<tr>
<td>Delafosse et al. 2008, RT, 150 rpm, 0.45 m, 0.15/0.45, 4, 0.15/0.45, T/10, T, water, 56,250</td>
<td>1,000,000 (full), hexahedrons, SM, CFD code: FLUENT 6.2.16</td>
<td>( \langle u \rangle )</td>
<td>Purpose: To compare URANS and LES RANS model: standard ( k-\varepsilon ) Notes: The URANS overestimated the width of the stream but gave correct order of magnitude in terms of turbulent dissipation rate in the whole vessel volume except in the vicinity of the impeller. Moreover, LES simulations predict the different contributions very well; however, the URANS simulation failed to predict the respective amounts of turbulent and periodic kinetic energy</td>
</tr>
<tr>
<td>Zadghaffari et al. 2010, 6-blade RT, 250 rpm, 30 cm, T/3, T/3, 4, 0.15, T, water, –</td>
<td>970,997 (full), hexahedral cell, SM, CFD code: ANSYS-FLUENT 6.3)</td>
<td>( \langle u \rangle )</td>
<td>Purpose: To compare URANS and LES RANS model: standard ( k-\varepsilon ) Notes: More reliable results of the mixing process were obtained by the LES compared with the RANS. A power number maximum deviation of 3% and ( \varepsilon ) of about 29% were underpredicted</td>
</tr>
</tbody>
</table>
4.3. Dispersed phase volume fraction

Another important factor in Eq. (5) is the dispersed phase volume fraction. This factor is defined by the momentum balance equation. Generally, knowledge on bubble size distribution in stirred vessels is necessary; hence, phase continuity, momentum equations, and population balance should be solved simultaneously. The multidimensional population balance model (PBM) accounts for these aspects simultaneously and can describe interactions between gas bubbles and continuous liquid both in terms of momentum and mass coupling. The first efforts in this field have been done by assuming monodisperse bubble size distribution during the investigation of different parameters in gas-liquid stirred systems (Khopkar et al. 2006, Scargiali et al. 2007); however, most of the authors focused on slip velocity in solid-liquid systems rather than gas-liquid systems.

The number density of bubbles of size $v$ at time $t$, $n(v,t)$, is introduced via the PBE:

$$\frac{\partial}{\partial t}n(v,t)+\frac{\partial}{\partial x}[\bar{u}_g(v,t)n(v,t)]=B_B-D_B+B_C-D_C$$

where $B_B$, $D_B$, $B_C$, and $D_C$ represent the birth rate due to breakup of larger bubbles, the death rate due to the breakup into smaller bubbles, the birth rate due to the coalescence of smaller bubbles, and the death rate due to the coalescence with other bubbles, respectively.

### 4.3.1. Bubble breakup and coalescence

There are many coalescence and breakage kernels developed for bubbly flow over the last 40 years, and they are written in similar forms except for some minor differences in assumptions or model constants. In a gas-liquid stirred Rushton vessel, bubble breakup occurs due to turbulent eddy collision and instability of large bubbles.

Bubble breakup occurs when disruptive forces in the liquid bulk are large enough to overpower bubble surface tension. Breakage frequency, daughter bubble size distribution, and size distribution of bubbles are the main components of the breakup kernel (Luo and Svendsen 1996, Lehr and Mewes 2001). Breakup rate depends on the operating parameters and fluid properties (both continuous and dispersed). Generally, there are four main mechanisms for deformation and breakup. They include turbulent fluctuation and collision, interfacial instability, viscous shear stress, and shearing-off process. Interfacial instability is due to density differences, which includes the Rayleigh-Taylor instability. The other three mechanisms are due to flow dynamic characteristics in the continuous phase. Breakup and deformation by viscous shear stress occur when the velocity gradient occurs across the interface (Liao and Lucas 2009). Meanwhile, shearing-off is an additional mechanism caused by velocity differences across the interface, which happens when fluid particle diameter increases and becomes unstable. The shearing-off mechanism is also called erosive breakage and is characterized by a number of small particles sheared off from a large one (Fu and Ishii 2003).

The most important mechanism for breakup and deformation is collisions with eddies or turbulent pressure fluctuations of the surrounding fluid. This breakup mechanism occurs when the dynamic pressure difference around the particle results in the oscillation amplitude. When the value reaches a critical point, the bubble surface deforms and stretches in one direction and finally breaks up into two or more daughter particles. Therefore, this mechanism can be characterized by a balance between dynamic pressure $\tau_d$ and its surface stress $\tau_s$. In stirred vessels, turbulent fluctuation is the most important factor for bubble collision and breakup.

The degree of breakup by turbulent fluctuation and collision can be classified by five main factors: (1) turbulent kinetic energy of bubbles greater than a critical value; (2) velocity fluctuation around bubble surfaces greater than a critical value; (3) turbulent kinetic energy of the hitting eddy greater than a critical value; (4) inertial force of the hitting eddy $F_e$; (5) a combination of criteria.

There is a variety of models based on these factors that can be used in CFD simulation. Prince and Blanch (1990) introduced a model that can predict bubble size distribution (as shown in Table 3). The model is based on the first mechanism, and the breakup rate is given by the collision rate of bubbles with turbulent eddies and breakup efficiency. Prince and Blanch (1990) also considered bubble coalescence due to laminar shear and different rise velocities. This model does not include daughter bubble size distribution after the breakage event of particle size. Lehr et al. (2002) further developed the Prince and Blanch model by combining the bubble coalescence due to different rise velocities and turbulent eddies. Laakkonen et al. (2007) later compared the two kernels and found that the Lehr model gives excellent prediction of local bubble size in bubble columns but underpredicts in gas-liquid stirred vessels.

The breakup model by Luo and Svendsen (1996) is also based on the first mechanism. It was developed for predicting fluid-particle breakup rates in a turbulent dispersion and assumes that bubble breakup happens when turbulent eddies hit the bubble surface. The model contains no adjustable or unknown parameters, so no prior assumption is required. The turbulent breakup mechanism can be modeled by the product of breakup probability due to the eddy energy contained and collision frequency between turbulent eddies and bubbles (Kerdouss et al. 2008).

The Luo and Svendsen (1996) model is more popular because it simultaneously investigates bubble breakup rate and daughter size distribution. Some researchers (Bordel et al. 2006, Podila et al. 2007) compared the various versions of the Prince and Blanch (1990) and the Luo and Svendsen (1996) models. They found that there is no great difference among gas holdup, mean flow, and bubble Sauter mean diameter ($d_{50}$) although they were predicted by means of different kernels. However, there are some differences in the predicted bubble size distribution, which depends on Sauter mean diameter. The results obtained by Luo and Svendsen (1996) were found to generate a real number of small bubbles as well as very large bubbles.

Bubble coalescence occurs when two or more bubbles collide and the film of liquid between them thins and ruptures. Collision rate and coalescence efficiency are components of the coalescence process. Generally, three mechanisms can be distinguished for the coalescence process: (1) film rupturing resulting in coalescence; (2) enough bubble contacting time for the small amount of liquid between them to become thinner than critical value of drainage; (3) the last but most important, collision of two bubbles and trapping of the liquid film between them.

Four criteria have been proposed for the coalescence process in a turbulent flow: turbulent fluctuations in the continuous phase, mean velocity gradients in the flow, interactions or helical/zigzag trajectories, and bubble capture in an eddy.

Several models are available for these components in the literature (Coulaloglou and Tavlarides 1977, Prince and...
### Table 3  Summary of breakup models.

<table>
<thead>
<tr>
<th>Breakup models</th>
<th>Remarks</th>
<th>Authors</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \Omega(V_i) = \sum_{k} c_i \left( d_i + \frac{2}{k} \right)^{3} \left( \frac{2}{k} \right)^{2/3} \left( \frac{2}{k} \right)^{2/3} )</td>
<td>1. Based on bubble collision with a turbulent eddy</td>
<td>Prince and Blanch 1990</td>
</tr>
<tr>
<td></td>
<td>2. Considering the breakup frequency by multiplying collision</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3. The collision frequency is assumed equal to the probability that turbulent eddies have sufficient energy to rupture a bubble</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4. Predicted the same and monotonic behavior even at high turbulent dissipation rates</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5. Problem with determining the integration limits</td>
<td></td>
</tr>
<tr>
<td></td>
<td>6. Daughter bubble size distribution based on uniform distribution model</td>
<td></td>
</tr>
<tr>
<td>( \Omega(V) = 0.92(1-\alpha) \left( \frac{a}{d} \right)^{3/6} \int \frac{1}{\min \left( \frac{1}{d}, \frac{1}{d} \right)} d \xi )</td>
<td>1. Based on bubble collision with a turbulent eddy</td>
<td>Luo and Svendsen 1996</td>
</tr>
<tr>
<td></td>
<td>2. Modified version of Prince and Blanch model by calculating the critical energy as the increase in surface energy during the breakage</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3. Most widely used model because it does not include any empirical or unknown parameter</td>
<td></td>
</tr>
<tr>
<td>( \Omega(V, V_i) = \int_{\min}^{\max} \frac{3.5 d_i (1-\alpha)}{d_i^{4/3} V_i^{4/3} \rho_i d_i^4} d \xi )</td>
<td>1. Based on the force balance</td>
<td>Lehr and Mewes 1999</td>
</tr>
<tr>
<td></td>
<td>2. The breakup probability depends on the angle the eddy hits the bubble</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3. Assumed that only eddies of length scale smaller than or equal to the bubble diameter can induce breakup</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4. The daughter bubble size distribution obtained directly from the breakup frequency</td>
<td></td>
</tr>
<tr>
<td>( \Omega(V, V_i) = 1.190 \left( \frac{a}{d_i} \right)^{3/6} \left( \frac{1}{d_i} \right)^{1/6} \min \left( \frac{1}{d_i}, \frac{1}{d_i} \right) )</td>
<td>1. Based on the force balance</td>
<td>Lehr et al. 2002</td>
</tr>
<tr>
<td></td>
<td>2. Modified version of (Lehr and Mewes 1999)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3. Not considered the angle under which the eddy hits the bubble</td>
<td></td>
</tr>
<tr>
<td></td>
<td>4. The breakup probability depends on the kinetic energy of the eddy exceeding the critical energy that is obtained from the force balance</td>
<td></td>
</tr>
</tbody>
</table>
gas molecules. This assumption is questionable because fluid bubble motion is interpreted similar to random motion of effective volume swept by moving bubbles per unit time. The coalescence time, coalescence occurs (Liu and Li 1999). In away or coalesce. If the random contact time exceeds the contact time when bubbles collide where they either bounce ruptures and bubbles coalesce.

Coulaloglou and Tavarides (1977) assumed that there is a contact time when bubbles collide where they either bounce away or coalesce. If the random contact time exceeds the coalescence time, coalescence occurs (Liu and Li 1999). In this model, the collision frequency is proposed by the effective volume swept by moving bubbles per unit time. The bubble motion is interpreted similar to random motion of gas molecules. This assumption is questionable because fluid particle collisions are neither elastic nor rigid. The relative velocity between bubbles is determined by assuming that colliding bubbles have the velocity of an equal-sized eddy and is determined by the mean square root of two equivalent eddy velocities. Inertial subrange of isotropic turbulence is always assumed for eddy velocity calculations. This model has some weaknesses arising from the assumption that all bubbles have the same velocity as equal-sized eddies and are in inertial subrange arbitrarily. As a result, some modifications have been made. The first modification was done by Colin and Riou (2004) in a gas-liquid system. He explained the effect of size ratio between eddies and bubbles and presented that if the integral length scale of turbulence $l$, smaller are than the particles, the relative bubble motion is mainly due to the mean shear of flow because turbulent eddies are not sufficient to move the particles. According to Colin and Riou (2004), turbulent collisions occur only when $(b/l; b < l)$ and $(b/l; b > l)$.

Other researchers modified the model for the second time (Hibiki and Ishii 2000a,b, Lehr et al. 2002) to investigate the increasing rate of particle collision frequency by increasing dispersed phase volume fraction. This factor has been considered by multiplying the original equation with a factor ($\gamma$). The third modification was to consider the ratio of the mean distance between particles to their average relative turbulent path length. According to Wu et al. (1998) and Wang et al. (2005), if the turbulent path length is smaller than the distance between particles, no collision should be counted. The effect can be described by multiplying the original collision frequency with a decreasing factor $\Pi$. (The coalescence equations and mentioned corrections are summarized in Tables 4 and 5, respectively.)

Hagesaether (2002) assumed that bubble coalescence depends on bubble size diameter, relative bubble velocity, and collision angle ($\beta$). In his model, coalescence decreases with the increase of bubble size diameter and efficiency decreases with increasing relative velocity. A critical relative velocity must exist for coalescence; in addition, coalescence efficiency is dependent on collision angle ($\beta$) in the moment of coalescence. Collision angle ($\beta$) is defined as the angle between different vector positions corresponding with colliding bubbles and the relative velocity vector (Hagesaether 2002). A complete review of methods for coalescence breakage problems is reported by Laakkonen et al. (2007).

### 4.3.2. Population balance solution method

Coupling PBE with CFD is another challenge in modeling multiphase flows. Bubble size equations formulation involves source terms for coalescence or breakup and transport by convection.

Generally, different techniques exist for solving PBEs: complete population balance model (PBM), bubble density model (BDM), methods of classes (CM), methods of moments (MOM), standard method of moments (SMM), the quadrature method of moments (QMMOM), direct quadrature method of moments (DQMMOM), conditional quadrature method of moments (CQMM), multiple size group model (MUSIG), and Monte Carlo method (MCM) are several numerical approaches for solving PBEs. These models (as shown in Table 6) provide a framework in which the PBM together with the coalescence and breakup models can be unified into 3D CFD calculations.

The BDM model is a simple model based on the calculation of gas-phase volume fraction by solving scalar type transport equations (Kerdouss et al. 2008). A coupled CFD-BDM

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**Table 4** Summary of coalescence models.

<table>
<thead>
<tr>
<th>Author</th>
<th>Coalescence rate</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>Prince and Blanch 1990</td>
<td>$C_{ij} = F_c (\theta_{ij}^{(1/3)} + \theta_{ij}^{(1/2)} + \theta_{ij}^{(2/3)}) \eta_0$</td>
<td>$\eta_0 = e^{(\delta_c/\lambda_t)}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$t_r = r_t / h_f$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\tau_0 = h_r / \ell_{ef}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$u_{rel} = (u_{ij}^c + u_{ij}^d)$</td>
</tr>
<tr>
<td>Coulaloglou and Tavarides 1977</td>
<td>$h(d_i,d_j) = c_i \pi d_i^{1/3} (d_i + d_j)^{1/3} (d_i^{1/3} + d_j^{1/3})^{1/3}$</td>
<td>$\lambda(d_i,d_j) = \exp \left[ \frac{-C}{\rho_c / \rho_L + 0.5} \left( \frac{d_i^{1/3} + d_j^{1/3}}{d_i^{1/3} + d_j^{1/3} + d_{ij}} \right) \right]$</td>
</tr>
<tr>
<td>Hagesaether 2002</td>
<td>$h(d_i,d_j) = c_i \pi d_i^{1/3} (d_i + d_j)^{1/3} (d_i^{1/3} + d_j^{1/3})^{1/3}$</td>
<td>$\lambda(d_i,d_j) = \exp \left[ \frac{-C}{\rho_c / \rho_L + 0.5} \left( \frac{d_i^{1/3} + d_j^{1/3}}{d_i^{1/3} + d_j^{1/3} + d_{ij}} \right) \right]$</td>
</tr>
</tbody>
</table>
has been used to predict the local bubble size by other authors (Lane et al. 2002, Kerdouss et al. 2006, Moilanen et al. 2008). In this approach, local bubble size varies in every single point in the region, but from point to point, a nonphysical monodisperse distribution is considered (Lane et al. 2002, Kerdouss et al. 2006, Moilanen et al. 2008), but its formulation suffers from several downsides: all equations related to bubble size have gathered together as a function of the critical energy dissipation rate and the Weber number without paying attention to the probability and rate of bubble-eddy and bubble-bubble collisions; proper bubble breakage and coalescence kernels are not included; the model is not fully predictive, although by adjusting some empirical constants introduced in the model, good predictions are accessible.

The CM is the most commonly used technique for solving PBE. In this method, the whole domain of internal coordinate space is subdivided into a finite set of bins. The PBE transforms from an integro-differential multidimensional equation into a set of differential equations of smaller dimension for the number of bubbles expected within each bin. The high computational demand is required to receive satisfactory prediction of the evolution of moments for the bubble size distribution is a shortcoming of MC.

MOM and SMM are techniques that require relatively low computational cost. System evolution is calculated through some lower-order moments of distribution using quadrature approximation. However, these methods suffer from some simplifications due to the closure problem (Rong et al. 2004). To overcome the closure problems, new methods were formulated based on the use of a quadrature approximation. McGraw (1997) have proposed the QMOM, which was validated and extended by Marchisio et al. (2003) using quadrature formula and solution of transport equation for lower-order moments of bubble size distribution. With QMOM, the transport equations for moments of number density function are solved by using quadrature approximations. QMOM is able to provide accurate predictions with relatively small numbers of quadrature points with reasonable computational time, although QMOM do have some limitations because only monovariate problems could be treated (Petitti et al. 2010).

Multivariate problems require alternative approaches such as DQMOM or CQMOM (Yuan and Fox 2011). With the former, each node of the quadrature can be treated as a distinct phase, with its own velocity, size, and composition. Also, weights and nodes of the quadrature approximation are calculated directly through transport equations in every point of the computational domain. DQMOM can also be easily combined with multiphase models. The DQMOM is another model that was reduced by Marchisio and Fox (2007) based on the QMOM by McGraw (1997). The main advantage of DQMOM is that only a few abscissas are necessary to describe a particle distribution because of the numerical quadrature approximation closure used (Selma et al. 2010). Recently, Buffo et al. (2012) has formulated an approach based on the DQMOM method to describe the evolution of the gas bubble in a realistic gas–liquid stirred vessel reactor. In their work, the initial conditions have been highlighted to obtain meaningful predictions and also a proper corrective term in all the tests conducted on a zero-dimensional homogeneous system (described by Marchisio and Fox 2005).

The MCM is another method that can accurately and realistically describe the behavior of a physical system. Each notional bubble is randomly selected, realizing a physical event artificially involving real bubbles. However, this method is not completely coupled with CFD for simulation of real systems because large numbers of notional bubbles considered are required to reduce the statistical error (Buffo et al. 2012).

The last approach to solve PBM is the MUSIG model, which is a framework wherein the PBM can be incorporated into 3D CFD calculations. With this technique, bubble size distributions are divided into a suitable number of size classes for which continuity equations are solved. Homogenous and inhomogeneous multiphase polydispersed
flows are two approaches of the MUSIG model. For inhomogeneous flows, the MUSIG model assumes that there are \( N \) different velocity groups that can be treated as a separate phase in a multiphase flow calculation. Each velocity group is then divided into different size groups, so \( N+1 \) sets of coupled equations (continuity and momentum) should be solved. In a homogenous flow, to decrease the large number of equations involved, MUSIG assumes that different size groups share the same velocity; thus, it is only necessary to solve one set of momentum equations for all bubbles, although continuity equations of the size groups are solved to present the size distribution (Moilanen et al. 2008, Montante et al. 2008).

### 5. Mass transfer

The aim of the models mentioned is to give a representative prediction of mass transfer, which is often controlled by the liquid side through the volumetric mass transfer coefficient \( k_{L,a} \). Based on the models mentioned above, several researches have focused on measuring mass transfer in stirred reactors containing gas and liquid. Table 7 summarizes the work on gas-liquid mass transfer prediction in stirred reactors via CFD simulations based on bubble size distribution.

The Luo and Svendsen model can be considered the most widely used model for the bubble breakup rate and the Prince and Blanch model is the most widely used for...
<table>
<thead>
<tr>
<th>Authors/year/system, impeller type, speed, T, D/T, C/T, N, W/T, Z/T, operating Re</th>
<th>Grid size, type, impeller motion model</th>
<th>Mass transfer model</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laakkonen et al. 2006, Rushton, 150–250 rpm, 630 mm, T/3, T/2, fully baffled, T/10, T, 155–250, water -</td>
<td>160,000, hexahedral (full), sliding, SM, CFD code: CFX-4.4</td>
<td>Two-film theory ( k_i = 0.301 \sqrt{D_i \left( \frac{\varepsilon}{\rho} \right)^{1/4}} )</td>
<td>Purpose: To find unknown parameters in bubble breakage, coalescence mechanistic and multicomponent gas-liquid mass transfer. Investigated parameters: bubble size distributions. Turbulence model: standard ( k-\varepsilon ) PB method: simplified discretized PB equation. Breakup model: Luo and Svendsen. Coalescence model: Coulaloglou and Tavlarides.</td>
</tr>
<tr>
<td>Kerouss et al. 2008, 45 pitched blade with 3 blades, 600 rpm, 12.5 cm, 0.6T, 2, 4/3T, water, ( 3.75 \times 10^4 )</td>
<td>1,000,000, hexahedral (full), MRF, CFD code: FLUENT 6.2</td>
<td>Penetration theory ( k_i = \frac{2}{\sqrt{\pi}} \sqrt{D_i \left( \frac{\varepsilon}{\rho} \right)^{1/4}} )</td>
<td>Purpose: To predict mass transfer. Investigated parameters: Sauter diameter, local mass transfer coefficient. Turbulence model: standard ( k-\varepsilon ) PB method: CM. Breakup model: Luo and Svendsen. Coalescence model: Hagesatther et al.</td>
</tr>
<tr>
<td>Moilanen et al. 2008, Rushton (RT), phase jet (PJ), and combi jet (CJ), 0.64 m, T/3, T/3, 4, T/10, T, water</td>
<td>&gt; 120,000 nodes, hexahedral (full), SM, CFD code: CFX 5.7.1</td>
<td>Correlation ( k_i = 0.3 \sqrt{D_i \left( \frac{\varepsilon}{\rho} \right)^{1/4}} )</td>
<td>Purpose: To investigate local variables. Investigated parameters: flow field comparison, volumetric density distribution, gas hold up, volumetric mass transfer rate. Turbulence model: SST. PB method: MUSIG, BND. Breakup model: Luo and Svendsen. Coalescence model: Coulaloglou and Tavlarides.</td>
</tr>
<tr>
<td>Xia et al. 2009, down-pumping propeller (DPP), 6-curved-blade disc turbine (6CBDT), 6-arrowy-blade disc turbine (6ABDT), 300~600, 0.3 (0.467, 0.043), 0.4, 0.3, 1.48</td>
<td>638,212, 744,157, 744,306, hexahedral (full), MRF, CFD code: ANSYS CFX 11.0</td>
<td>Based on Higbie theory ( k_i = \frac{2}{\sqrt{\pi}} \sqrt{D_i \left( \frac{2p}{\rho} \right)^{1/2}} )</td>
<td>Purpose: To investigate fluid dynamics in three bioreactors equipped with different impeller combinations. Investigated parameters: velocity profiles, average shear stress in the bulk, oxygen mass transfer coefficient, material concentration. Turbulence model: multiphase ( k-\varepsilon ) PB method: QMOM. Breakup model: Prince and Blanch. Coalescence model: Prince and Blanch.</td>
</tr>
<tr>
<td>Selma et al. 2010, a double 6-bladed standard Rushton impellers, 450, 0.292, T/3, T/10, T/2, 1.5T, 2T, 7.1x10^4</td>
<td>255,000 cells, hexahedral (full), MRF, CFD code: open FOAM</td>
<td>Eddy cell ( k_i = 0.4 \left( \frac{\varepsilon}{\rho} \right)^{1/4} \left( \frac{D_i}{\varepsilon} \right)^{1/2} )</td>
<td>Purpose: To validate the DQMOM with the CM. Investigated parameters: Sauter mean diameter, liquid velocity profile, gas volume profile. Turbulence model: standard ( k-\varepsilon ). PB method: DQMOM, BND. Breakup model: Luo and Svendsen. Coalescence model: Prince and Blanch.</td>
</tr>
<tr>
<td>Ranganathan and Sivaraman 2011, dual standard Rushton turbines, 450, 0.292 m, 0.3, 0.5, 2, 0.1, 2, water</td>
<td>Hexahedral (full), MRF, CFD code: ANSYS CFX11</td>
<td>Penetration theory, slip velocity, eddy cell, and rigid theory ( k_i^{slip-velocity} = \frac{2}{\sqrt{\pi}} \sqrt{D_i \left( \frac{v_i}{d_i} \right)^{1/2}} )</td>
<td>Purpose: To investigate hydrodynamics and mass transfer. Investigated parameters: axial and radial gas holdup, Sauter mean bubble diameter, local specific area. Turbulence model: standard ( k-\varepsilon ). PB method: MUSIG. Breakup model: Luo and Svendsen. Coalescence model: Prince and Blanch.</td>
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</table>
the coalescence rate. In the work of Sivaraman, hydrodynamics and mass transfer have been simultaneously taken into account to predict the mass transfer in gas-liquid stirred vessels (Ranganathan and Sivaraman 2011). Oxygen transport equations and hydrodynamic simulation has been individually done in other works. For a good prediction of mass transfer in these systems, the gas-liquid mass transfer, PBM, and CFD should incorporate to each other. Laakkonen et al. (2006) obtained a multiblock model for calculating mass transfer area by fitting unknown parameters in the bubble breakage and coalescence models against local experimental data. The fitting included simulations along with sensitivity and correlation analysis to reduce parameter space. They have reported that fitted models are useful tools for scale-up and reactor design.

Kerdouss et al. (2008) used converged steady-state single primary phase flow field by MRF as an initial condition for the transient two-phase calculations to avoid numerical difficulties. A single bubble size was used as the initial condition with PBEs to improve the overall speed of the calculations.

Moilanen et al. (2008) simulated the effect of impeller geometry on G-L mass transfer with two different PB approaches (MUSIG and BND) and two equation turbulence models (k-ε and k-ω, SST). The BND and MUSIG models predicted different trends for vessel-averaged bubble size using different impellers; but unfortunately, the authors did not justify the reasons, but they explained that the BND approach is a useful and fast design tool even if it lacks the local BSD. Generally, a good prediction of estimated local bubble size was exhibited, based on fitted model parameters in the breakage and coalescence terms from the multiblock study.

Gimbun et al. (2009) have focused on the development of a modeling approach for gas-liquid stirred vessels. First, they performed constant bubble size with the spherical shapes along with the Schiller and Naumann (1935) drag model with PBM; next they used a non-uniform bubble size with two different drag models: the hard sphere drag model of Schiller and Naumann (1935) and Ishii and Zuber (1979). They have reported that by means of a nonspherical drag model (CFD-PBM-IZ), the results improved significantly because of the fact that the effect of local bubble sizes on the two phase flow is mainly via the interphase exchange coefficient, which depends on the drag model.

Xia et al. (2009) have investigated different fluid dynamics of a bioreactor equipped with different impeller combinations CFD simulation was used for validation of their work and finding optimum situations of vessel domain that affect the fermentation process.

Selma et al. (2010) have compared the DQ MOM and the CM in a rectangular bubble column and stirred vessel reactor. Both methods are in agreement with experimental results, although DQ MOM is computationally more efficient to use in comparison with CM. The authors reported that using CM, both the 15 and the 25 classes have produced reasonable agreement with experimental data, but the 25 classes appeared more accurate.

Ranganathan and Sivaraman (2011) used both inhomogeneous and homogenous MUSIG equations to account for the bubble size distribution and mass transfer. The polydispersed gas phase was subdivided into 21 groups ranging from 0.1 to 10 mm for the homogenous and two velocity groups (Air1: between 0.1 and 5 mm; Air2: between 5 and 10 mm) for the polydispersed gas phase for the inhomogeneous MUSIG equations. However, Air2 showed higher discrepancy in the results of the local Sauter mean bubble diameter compared to the velocity group Air1 because the bubble size in the reactor is not in the range of 5–10 mm. In addition, they applied different mass transfer coefficients based on various approaches such as penetration theory, eddy cell model, slip velocity model, and rigid-based model. They reported that the mass transfer coefficient predicted by the penetration theory was higher than the eddy-cell model value predicted by the penetration theory. Both these models have overpredicted the value of the mass transfer coefficient in the impeller discharge depending on the eddy dissipation rate. The slip velocity model and the eddy cell model closely matches with experimental data.

Generally, bubbles in stagnant zones accumulate and coalesce and create bubbles with larger diameters; meanwhile, bubbles break up in the vicinity of the impeller due to the high shear and the BSD is shifted to lower bubble diameters. Every single bubble has its own velocity, size, and composition, which must be considered to correctly predict the evolution in time and space for local and global mass transfer flux predictions. So, by coupling accurate PBM with CFD, this aim will be accessible. Unfortunately, no studies have considered all these parameters together and instead used simple mass transfer coefficient models for prediction of gas-liquid mass transfer.

### 6. Impeller motion model

Subsequent to selecting a suitable equation, suitable boundary conditions for impeller blades and disc surfaces pose a challenge because simulation of these moving sections is attached to both stationary baffles and impeller. There are several techniques available to address this issue. Using the impeller boundary conditions (IBC) method is the simplest strategy, which imposes suitable turbulence and velocity quantities on the surface swept by impeller blades. However, this method requires the availability of experimental data. Other fully predictive procedures are available, with no requirement for experimental data as boundary or initial conditions (Montante et al. 2001). Sliding mesh (SM), multiple reference frame (MRF), and inner-outer (IO) are three approaches for the simulation of flow field in modeling problems that involve both stationary and moving zones (Ankamma Rao and Sivashanmugam 2010). The first is based on transient computation to produce time-accurate flow field, and the other two are based on steady-state computation producing time-average flow field, resulting in considerable savings of computational demand compared with the SM approach (Ankamma Rao et al. 2010).
and Sivashanmugam 2010). The choice of grid characteristics and computational domain is essential because it may strongly affect the quality of results, especially in the MRF framework. The coupling between both domains must be taken into account because if the mesh is not fine enough, there will be a gap between the small cells around the impeller and the large cells at the vessel. In this case, both grids will not coincide on the surface at the interface resulting in a high numerical discretization error (Abu-Farah et al. 2010). Thus, this method is appropriate when flows at the boundary between the inner and the outer zones are nearly uniform. This approach also cannot predict the rate of decay in the local maximum velocity in the wall jet (Ochieng et al. 2009). For more details, refer to Joshi et al. (2011a).

7. Discretization schemes and equation solver algorithms

Important factors that affect accuracy and precision of flow simulations in stirred vessels are discretization schemes and equation solver algorithms. Choices are constrained by the computational power available and the basis of dominating transport phenomena; in other words, selected solution algorithm should be physically realistic and satisfy overall balance (conservative). Four basic rules should be satisfied: when a face is common to two adjacent control volumes, flux across it must be represented by the same expression in discretization equations for both the control volumes; all coefficients in governing the differential equation must always be of the same sign; the slope linearization of source term should be negative so erroneous results must always be of the same sign; and equation solver algorithms should satisfy the governing differential equation (see Table 8).

Central differencing scheme (CDS), upwind differencing scheme (UDS), power law, central differencing, hybrid differencing scheme (HDS), and quadratic upstream interpolation for convective kinetics (QUICK) are the widely used discretization schemes. The CDS does not recognize strength of convection or direction of flow for accuracy and stability. Either grid spacing should be small or velocity should be very low (Pe<2). The UDS is a first-order discretization scheme used for initiating a simulation and is desirable for stable iteration because the direction of flow is inbuilt in the formulation. The HDS is based on the ratio of the convective flow to diffusion (Peclet number) and depends on the flow and fluid properties. In other words, the HDS is a combination of the CDS and the UDS. It is identical to the CDS for Pe between -2 and 2, and outside this range, it reduces to the UDS. The Power law scheme is also similar to the HDS, but it is more accurate.

Discretization schemes have been dealt with in very few works. Sahu et al. (2005) compared the upwind, power law, and hybrid as three first-order numerical schemes. They explained that the first two give similar results and converge more quickly, whereas the solution of upwind scheme was more robust. For discretizations that are first order in terms of Taylor series, a truncation error can be minimized by using higher-order schemes. Higher-order discretization involves more neighbor points and reduces discretization errors by bringing wider influence. Brucato et al. (1998) compared the results of the hybrid upwind scheme and the third-order upwind discretization scheme, QUICK, and claimed that the only difference was QUICK tends to predict a little higher recirculation rates in the bottom and top of the tank. Roache (1998) concluded that the effect of inherent numerical diffusion of the first-order methods on solution accuracy is devastating. Aubin et al. (2004) also indicated this result.

8. Conclusions and challenges in CFD simulations of stirred vessels

Generally, the results of this review have been briefed in the following:

- The E-L viewpoint can simulate detailed flow structures with a higher spatial resolution than E-E and shows more benefits for dispersed multiphase flows but needs more computational cost.
- Generally, the LES method has obtained better results in comparison with the other approaches especially with the RANS method, which, with respect to the measured mixing time, the simulated one has been overestimated in most some cases. The major weakness of the k-ε model is the assumption of isotropy of turbulence in the vessel, which results in less accurate predictions in regions of anisotropic turbulence, despite the generally satisfactory comparison between the experimental findings and the numerical predictions, especially in regions far away from the impeller (Markatos 1986).
- There are still difficulties related to turbulence models in terms of flow data such as mixing time scales to

<table>
<thead>
<tr>
<th>Scheme</th>
<th>Conservative</th>
<th>Bounded</th>
<th>Accuracy</th>
<th>Transportive</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>CDS</td>
<td>Yes</td>
<td>Conditionally bounded</td>
<td>Second order</td>
<td>No</td>
<td>Unrealistic solutions at large Peclet number</td>
</tr>
<tr>
<td>UDS, HDS, PLS</td>
<td>Yes</td>
<td>Unconditionally bounded</td>
<td>First order</td>
<td>Yes</td>
<td>Induce false diffusion if the velocity vector is not parallel to one of the coordinate directions</td>
</tr>
<tr>
<td>QUICK</td>
<td>Yes</td>
<td>Unconditionally bounded</td>
<td>Third order</td>
<td>Yes</td>
<td>Less computationally stable</td>
</tr>
</tbody>
</table>

Can give small overshoot and undershoot
device performance. This aspect has not been thoroughly considered by researchers. CFD still poses a problem in processes that involves significant flow behavior changes, phase change, and combining of mixing where the fluid properties change during the reaction within the reactor (such as polymerization).

- Due to the complexity of the flow field in stirrer reactors, the current available breakup and coalescence models for gas-liquid flows in these vessels are not adequate for many practical applications. Bubbles or droplets are typically polydispersed and are not necessarily spherical, which changes the active surface and affect mass transfer. Also, every single bubble has its own velocity, size, and composition, and all these issues must be considered along with the interaction between the phases to predict the evolution in time and space of the dispersed phase correctly that results in the correct local and global mass transfer flux predictions.

- For accurate prediction of mass transfer in stirrer vessels, both the hydrodynamic parameters and mass transfer parameters should be considered; thus, coupling of PBEs along with the bubble breakup and coalescence and CFD is necessary. Considering the issues mentioned, further work based on CFD must be done to better understand the relationship between flow field and the other parameters in the mixing reactors.

- CFD development is still a challenge, particularly with industrial-grade meshes and large hydroturbine simulation problems. A proper interpolation algorithm to correctly evaluate the fields is still missing. This is especially true in the presence of meshes where the size of cells on either side of the mixing plane varies significantly, or rather, where a “grid-independent” solution is required. Generally, CFD development has two steps: first, make it run, then make it run fast. It is at the “first make it run” stage of development that there are still many aspects left to uncover.

**Nomenclature**

- **Ar** Archimedes number $\left( Ar = g d_b^4 / \nu^2 \left[ \left( \rho - \rho_i \right) / \rho_i \right] \right)$
- **c** Constant (value = 0.6); this model is referred as the rigid model and is denoted as $k_{rigid}$
- **$D_L$** Diffusion coefficient
- **d** Diameter of parent bubble, m
- **$d_e$** Eddy size, m
- **$d_d$** Diameter of daughter bubble, m
- **$d_b$** Bubble diameter
- **$E$** Cumulative dissipated energy, m² s⁻¹
- **$F_{w,b}$** Volume fraction, $V_b / V$
- **F** Cumulative $\chi^2$ distribution
- **g** Gravitational acceleration, m s⁻²
- **h** Collision frequency, m³ s⁻¹
- **k** Wave number of eddies, m⁻¹
- **K** Von Karman constant
- **$L_m$** Mixing length (LES)
- **m** Number of daughter bubbles
- **$n_i$** Volume density distribution of energy dissipation rate
- **n** Number of bubbles or eddies, m⁻¹
- **N** Average number of eddies that arrive at the surface of a bubble in unit time
- **p** Viscosity ratio
- **$p_b$** Breakup efficiency
- **$Q_i$** Exchange flow rate between impeller and circulation zones, m³ s⁻¹
- **$Q_o$** Exchange flow rate between baffle and circulation zones, m³ s⁻¹
- **$S_h$** and **$S_c$** Source terms for number density generated by coalescence and breakup, respectively
- **u** Velocity vector
- **$\bar{u}$** Filtered or solved instantaneous velocity (LES)
- **$u_i$** Instantaneous velocity
- **$\tilde{u}$** Periodic velocity
- **$u'_i$** Turbulent velocity
- **$\overline{u'_i}$** Mean velocity solved by URANS simulation for the blade position $p$
- **$\langle u'_i \rangle$** Phase average velocity for the blade position $P$
- **$u_{s}$** Slip velocity
- **$\langle u_i \rangle$** Time average radial velocity, m s⁻¹
- **$\langle u_t \rangle$** Time average tangential velocity, m s⁻¹
- **$\langle u_r \rangle$** Time average axial velocity, m s⁻¹
- **We** Weber number

**Greek symbols**

- **$\alpha$** Volume fraction of disperse phase
- **$\beta$** Smagorinsky eddy viscosity ($\beta = 2/9$)
- **$\bar{\beta}$** Daughter size distribution
- **$\epsilon$** Turbulent dissipation rate, m² s⁻¹
- **$\nu_b$** Bubble slip velocity
- **ν′** Cinematic viscosity
- **$\nu_S^{SG}$** Subgrid turbulent viscosity (LES)
- **ν₁** Dynamic viscosity, kg m⁻¹ s⁻¹
- **$\nu^{SG}$** Subgrid turbulent stress tensor (LES)
- **μ** Dynamic viscosity, Pa s
- **$\mu_b$** Impeller volume ratio
- **$\mu_w$** Baffle volume ratio
- **$\mu'$** Turbulent viscosity, kg m⁻³ s⁻¹
- **$\mu_s$** Surface tension, N m⁻¹
- **$\nu$** Kinematic viscosity, m² s⁻¹
- **$\sigma$** Inertial turbulent stress, N m⁻²
- **$\Omega$** Breakup frequency, s⁻¹
- **$\Omega_b$** Source term for interfacial area density generated by surface instability, m⁻¹ s⁻¹

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